SPECTRAL ANALYSIS OF RADIOXENON

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ABSTRACT

Monitoring changes in atmospheric radioxenon concentrations is a major tool in the detection of an underground nuclear explosion. Ground-based systems like the Automated Radioxenon Sampler /Analyzer (ARSA), the Swedish Unattended Noble Gas Analyzer (SAUNA) and the Automatic portable radiometer of isotopes Xe (ARIX), can collect and detect several radioxenon isotopes by processing and transferring samples into a high-efficiency β-γ coincidence detector. The high-efficiency β-γ coincidence detector makes these systems highly sensitive to the radioxenon isotopes ^{133}Xe, ^{131m}Xe, ^{133m}Xe, and ^{135}Xe.

The standard analysis of the 2-dimensional beta-gamma energy spectra uses regions of interest (ROI) to determine the amount of a particular radioxenon isotope present by summing the counts in region. This method relies on the peaks of interest falling within energy limits of the region. Some potential problems inherent in this method are the reliance on stable detector gains and a fixed resolution for each energy peak. In addition, when a high activity sample is measured, there will be more interference among the ROI, in particular within the ^{133}Xe, ^{133m}Xe, and ^{131m}Xe regions.

A solution to some of these problems can be obtained through spectral fitting of the data. Spectral fitting is simply the fitting of the peaks using known functions to determine the number and relative peak positions and widths. By knowing this information it is possible to determine which isotopes are present. The area under each peak can then be used to determine an overall concentration for each isotope. Using the areas of the peaks, several key detector characteristics can be determined: efficiency, energy calibration, energy resolution, and ratios between interfering isotopes (Radon daughters).

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OBJECTIVES

Monitoring radioactive releases from nuclear explosions is a major component of the International Monitoring System (IMS) network (Hayes 1999, Bowyer 1999). Systems such as ARSA, SAUNA, and ARIX collect and detect several radioxenon isotopes by processing and transferring samples into a high-efficiency β-γ coincidence detector for analyses in support of treaty verification. The high efficiency β-γ coincidence detector makes these systems highly sensitive to the radioxenon isotopes ^{133}Xe, ^{131m}Xe, ^{133m}Xe and ^{135}Xe.

Standard radioxenon analysis uses ROIs in the beta-gamma energy correlation spectrum to determine radioxenon isotopic concentrations present. The ROI method relies on setting region energy limits such that the peaks of interest are fully encompassed. Some potential problems inherent in this method are the reliance on stable detector gains and a fixed resolution for each energy peak. Currently, the ARSA system uses 7 ROIs (Figure 1) while the SAUNA system uses 10 ROIs.

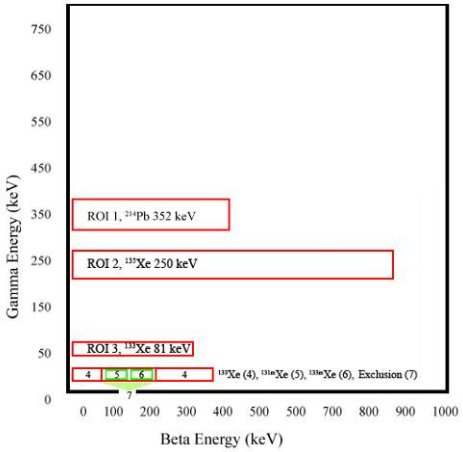

Figure 1. Standard ARSA 7 ROI.

The objective for spectral fitting is to supplement and/or replace the ROI beta-gamma energy correlation spectrum analysis method by fitting functions to the data. Several advantages are realized by using spectral fitting. First, detector gains and energy resolution can be checked by using an appropriate fitting algorithm, peaks can be found by differentiating the spectrum and looking for zero crossings. By performing a Gaussian fit to a peak, the centroid, sigma width, and pulse height are determined. The pulse height and width are used to calculate the peak area. The centroid of each peak is used for isotopic identification and to check for detector gain shifts. Changes in peak widths can indicate potential damage to a detector. A second advantage of spectral fitting is a reduced reliance on detector background measurements due to the ability to do a spectral background subtraction (often a linear fit). This in turn will reduce system setup time. Finally, an automated detector efficiency and energy calibration measurement can be made using spectral analysis. An automated method uses an algorithm to determine the peak position and then performs Gaussian fits to determine peak area and channel.

RESEARCH ACCOMPLISHED

Fielded radioxenon systems have traditionally used the ROI beta-gamma energy correlation spectrum analysis method to make xenon concentration measurements. Pacific Northwest National Laboratory (PNNL) has made progress towards switching to a spectral analysis method instead. Although fielding a system using spectral analysis is still a considerable time away, a first attempt at an automated spectral fitting algorithm has been made based on the Cern ROOT data analysis framework (http://root.cern.ch/). However, for spectral fitting to be most effectively used, a nuclear physicist should perform and check the fits rather than rely on an automated algorithm. An automated fitting routine has been done at PNNL.

Energy Calibration

One of the first attempts to do spectral fitting at PNNL was in characterizing the beta-gamma coincidence detector. The first step in characterizing a detector is to determine the energy calibration. This is done by fitting multiple energy peaks and correlating the channel of the peak centroid with the corresponding energy from the literature. For a beta-gamma detector, the gamma energy calibration is initially determined using a ^{137}Cs source, and a radioactive gas spike of ^{222}Rn and ^{131m}Xe. These sources give seven gamma- and x-ray energies ranging from 30 keV up to 662 keV. The seven gamma- and x-rays cover a good range of energies for the purpose of the detector.

A first step in performing a spectral fit is to subtract the background events. Accounting for background events is normally done by subtracting a background spectrum; however, it is also possible to fit a polynomial (Figure 2) to the sections of the spectrum that do not contain peaks. One assumption made when fitting the background is that there are no discontinuities present in the spectrum. By using a fit to subtract the background, measurement times are significantly reduced without a reduction in counting statistics.

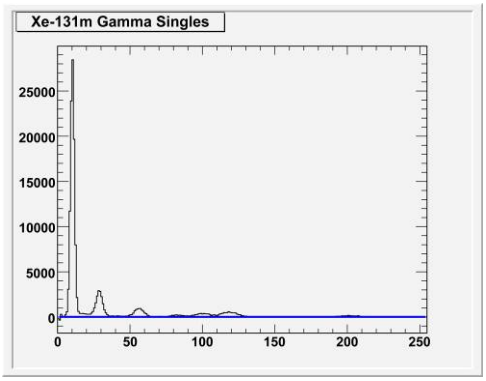

Figure 2. A ^{131m}Xe spectrum with both Radon daughters and ^{133}Xe present, where the blue line is the linear background fit.

Although background subtraction is not strictly necessary for determining the energy calibration and energy resolution of a detector, it is still best practice. Once the background is subtracted, a fitting algorithm can be run which performs Gaussian fits to those peaks present (Figure 3). Several limits are placed on the fitting algorithms to ensure a good fit. One limit is set based on the relative differences in expected energies. For example, ^{133}Xe has a 30 keV X-ray and 80 keV gamma-ray so the peak position should reflect that relative difference. Other limits might include setting a range on the Gaussian peak area relative to the literature-listed intensities, the number of Gaussian fits, and Gaussian widths. Knowledge of general detector characteristics is invaluable when limiting fitting algorithms. For example, the energy resolution, or full width half maximum of the peak, as a function of energy is proportional to $1/\sqrt{E}$.

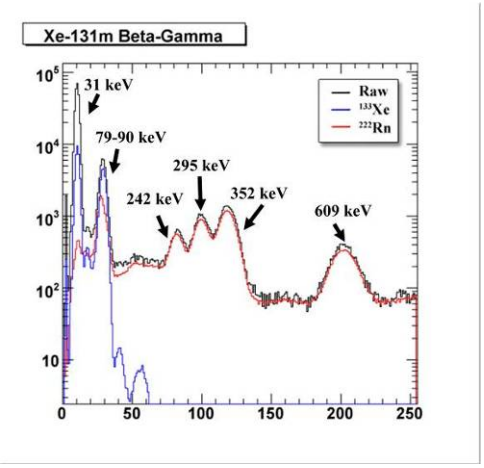

Figure 3. A ^{131m}Xe spectrum with multiple-fitting algorithms performed. Blue: ^{133}Xe peak fits and in Red: ^{222}Rn daughter peak fits.

Once the energy calibration for the gamma-ray detector has been established, the energy calibration for the beta detector needs to be determined. Using a ^{137}Cs source inserted along the beta detector it, becomes a relatively simple matter to measure the energy calibration. The 662 keV gamma-ray Compton scatters to produce a beta-gamma coincidence, which can be used to determine the beta-energy calibration (Figure 4). By fitting the diagonal line of constant energy (662 keV), a correlation between the gamma-energy calibration and beta-energy calibration can be defined, which in turn will determine the beta energy calibration. The energy calibration should be verified using the ^{131m}Xe peak which has a 30 keV x-ray in coincidence with a 129 keV conversion electron (CE), giving a coincidence peak of known energy (Figure 5).

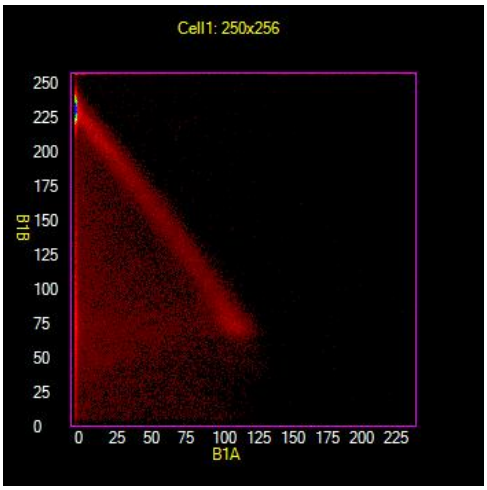

Figure 4. Beta-gamma coincidence ^{137}Cs spectrum.

Efficiency Measurement

Once the detector energy calibration is known, the detector efficiency is determined. The detection efficiency for a given detector is the number of detected events divided by the number of total events. A general formula used in calculating the efficiency is shown in Equation 1:

$$\text{Number of Total Events} = \frac{Counts}{\varepsilon \cdot Br} \quad \textbf{(1)}$$

$\varepsilon = \text{efficiency}$

Where, $Br = \text{branching ratio}$

$Counts = \text{Number of counts in peak of interest}$

The first step is to use the ^{131m}Xe data. By comparing the number of events detected in beta singles versus the number of events detected in beta-gamma coincidence, the gamma efficiency can be determined from Equation 2.

$$\frac{Counts_{\beta\gamma}}{\varepsilon_\gamma' \varepsilon_\beta' Br'} = \frac{Counts_\beta}{\varepsilon_\beta Br} \Rightarrow \varepsilon_\gamma' = \frac{Counts_{\beta\gamma}}{Counts_\beta} \quad \textbf{(2)}$$

In equation 2 the branching ratios are the same since there will always be an x-ray emitted in coincidence with the CE. The calculation also assumes ~100% detection efficiency of the 129 keV CE (partially due to an assumed low self attenuation) and a large detection solid angle (~ 99%). The 30 keV gamma-ray efficiency is then used in determining the beta efficiency for ^{133}Xe. Using Equation 3, the ratio between the gamma singles and beta-gamma coincidence will yield the efficiency for the betas in coincidence with the 30 keV x-ray for ^{133}Xe. Assuming this beta efficiency is the same as that for betas in coincidence with the 80 keV gamma-ray from ^{133}Xe, it is possible to

determine the gamma efficiency at 80 keV. By using data collected from the Radon daughters (^{214}Pb) it is possible to determine the gamma detection efficiencies at 242, 295, and 352 keV.

$$\frac{Counts_{\beta\gamma}}{\varepsilon_\gamma' \varepsilon_\beta' Br'} = \frac{Counts_\gamma}{\varepsilon_\gamma Br} \Rightarrow \varepsilon_\beta' = \frac{Counts_{\beta\gamma} Br}{Counts_\gamma Br'} \quad \textbf{(3)}$$

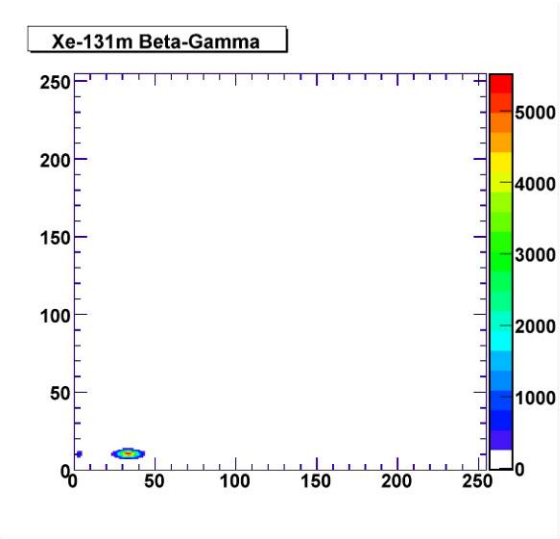

Figure 5. Beta-gamma coincidence spectrum used for determining the gamma-detection efficiency at 30 keV.

Deconvolution

Spectral fitting can also be used to perform deconvolutions. When multiple gamma rays are detected near the same energy, the resulting peak may fall within a single beta-gamma energy correlation spectrum ROI. However, when performing spectral fitting, it is possible to fit an asymmetrical peak with multiple Gaussian functions to resolve a more complex peak structure. This technique is often challenging and relies on knowledge of the nuclear structure for the isotopes detected. The majority of the nuclear structure is from the literature and can be imbedded into the isotopic identification and calculation. Several methods that can be used to determine if a peak is a multiplet, ranging from a wider peak than expected to actual peak structure (a shoulder on a peak). Once a multiplet peak has been identified, the number of Gaussian peaks used should be determined by matching the Gaussian width with the expected energy resolution at the particular energy (too many Gaussians will give a smaller width, while too few will give a wider width). Furthermore, the sum of the Gaussians should fit the overall peak of interest. Deconvolving a peak using spectral fitting is much easier to do in a manual fashion than automatically and is one of the reasons for spectral fitting being a supplement to the standard energy spectrum ROI method.

Fermi-Kurie plot

Given the difficulty in fitting a beta continuum, it is important to find an alternative method. A Fermi-Kurie plot (Krane 1988) is one method, which allows a beta spectrum to be plotted as a line. By plotting

$$\sqrt{\frac{n(T)}{\sqrt{T^2 + 2mc^2 T(T + mc^2)}}} \text{ versus T,} \quad \textbf{(4)}$$

Where, $n(T) = \text{number of counts}$

$T = \text{energy - rest mass}$

$mc^2 = 511\,\text{keV}$

a nearly straight line is plotted which crosses the x-axis at the end-point energy for the beta decay. There is a corrective term (Fermi function) that has been neglected which corrects for low energy non-linearities. The corrective function takes into account the initial and final spin and polarity states.

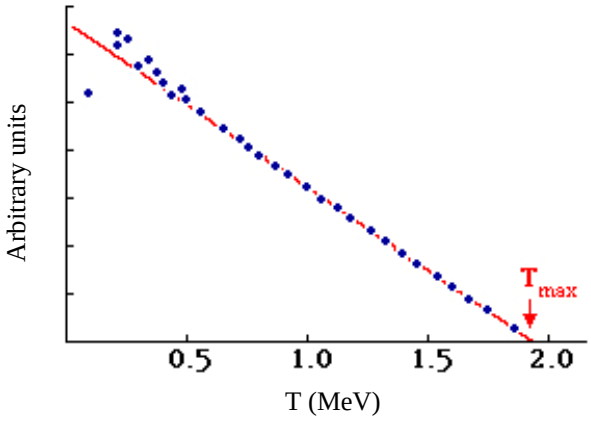

Figure 6. Fermi-Kurie plot.

CONCLUSIONS AND RECOMMENDATIONS

Spectral fitting is a supplement and potentially a replacement for the standard energy spectrum ROI analysis technique for radioxenon measurements. The primary function used in spectral fitting is combining Gaussian and polynomial functions to fit an entire spectrum. The fitting is slightly more complex for a beta continuum given the smooth curve of the spectrum, but by using a Fermi-Kurie plot, the beta spectrum becomes linear and is easier to fit. Given the number of peaks, relative positions, widths, and end-point energies, it is possible to uniquely identify the isotopes present, number of decays detected, and thus the concentration for each isotope.

Although spectral analysis poses many advantages, additional work needs to be completed to successfully convert current analysis techniques to spectral fitting. Spectral analysis works best given good statistical data. Without relatively high statistics, the fitting algorithm begins to fail. This is an area that can be addressed by doing the fits manually; however, it would be best to have an algorithm do it automatically. The other area that should be addressed is to make multi-dimensional fitting algorithms (fit the beta-gamma coincidence data rather than the projected data). Multi-dimensional fitting would make the best use of the data collected and could be more effective at removing interference terms.

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